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Project Summary

Support Studies in Fluidized-Bed Combustion—Annual Report, October 1978-September 1979

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This work supports the development studies for atmospheric and pressurized fluidized-bed coal combustion. The work was funded by the U.S. Environmental Protection Agency and the U.S. Department of Energy. Laboratory and process development studies are aimed at providing needed information on limestone utilization, removal of particulates and alkali metal compounds from the flue gas, control of SO_2 and trace pollutant emissions, the mechanism of attrition in fluidized-beds, a method of determining the attrition resistance of sorbents, and other aspects of fluidized-bed coal combustion. This report, upon which this summary is based, is available from NTIS (see box on last page).

Introduction

Hot Gas Cleanup

Removal of Alkali Metal Compounds from Hot Flue Gas—The use of diatomaceous earth and activated bauxite as solid granular sorbents for the removal of gaseous alkali metal compounds (*viz.*, NaCl, KCl, or K_2SO_4) from the hot combustion gas of coal continues to be studied. In these studies, a hot (800°C) simulated flue gas of PFBC loaded with a gaseous alkali metal compound passes through a hot packed bed of the granular sorbent being tested to determine the effectiveness of the sorbent for removing the alkali vapors.

Experimental tests have been made (1) to determine the effect of NaCl vapor concentration in the flue gas on the sorption performance and ultimate alkali sorption capacities of diatomaceous earth and activated bauxite and (2) to investigate the regenerability of activated bauxite by water-leaching. The average rate of NaCl vapor sorption for both sorbents was found to be a linear function of NaCl vapor concentration in the flue gas. Reduction in NaCl vapor concentration did not reduce the sorption efficiencies of either sorbent. The ultimate sorption capacity of activated bauxite was found to be about 25 mg NaCl/g sorbent, while the ultimate sorption capacity of diatomaceous earth was much greater but was not measured.

Experimental evidence indicates that the sorption of NaCl vapor by diatomaceous earth is due to the chemical reaction of diatomaceous earth with NaCl vapor to form water-insoluble sodium silicates. In contrast, activated bauxite captures NaCl vapor mainly through an adsorption mechanism.

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Since an adsorption process is a reversible process, activated bauxite is regenerable. Tests have demonstrated that activated bauxite can be easily regenerated by a simple water-leaching process and that the regenerated activated bauxite maintains a sorption activity similar to that of fresh activated bauxite.

Cost evaluations have been made as preliminary estimates of the costs of diatomaceous earth and activated bauxite used as granular sorbents for the removal of alkali vapors from the hot flue gas of PFBC's. Results indicate that on a once-through basis, the cost of activated bauxite is about twice that of diatomaceous earth; however, the cost of activated bauxite can be significantly reduced if calculated on the basis of regeneration.

Particle Removal from Flue Gas

In PFBC, the hot flue gas from the combustor must be expanded through a gas turbine to recover energy and make the process economical. To prevent unacceptable erosion of the turbine blades by particles of limestone and fly ash present in the flue gas, the particle loading must be reduced to low, but not well-defined levels. There is considerable concern that existing particulate control technology may be inadequate at the high-temperature, high-pressure environment of the PFBC flue gas. This report presents (1) a summary of the progress which was made in developing a pulse-jet resonant manifold system for investigating acoustic dust conditioning as a preconditioning step in particulate removal and (2) the results of testing a high-efficiency cyclone.

Acoustic Dust Conditioning—Work was continued at the University of Toronto, under subcontract to ANL, to develop a pulse-jet acoustic dust conditioning system for subsequent testing and evaluation at ANL. Acoustic dust conditioning is defined as the process whereby the natural tendency of particles to agglomerate is enhanced by exposure of the particles to high-intensity acoustic fields. The effort at the University of Toronto was scheduled to be completed in three phases. Phase I was completed; it consisted of the design, fabrication, and testing of a pulse-jet resonant manifold system (PJ-RMS) at atmospheric pressure. Sound intensities were as high as 160 dB at frequencies of 300 to 350 Hz.

Phase II, increasing the operating pressure of the PJ-RMS to about 1.01 MPa, was in progress when the project was discontinued due to budgetary factors. During Phase II, the operating pressue of the pulse-jet had been increased to 350 kPa, with indications that the increase in pressure had increased the sound levels obtainable.

Phase III, which was to be the design, manufacture, and testing of pulse-jet acoustical agglomeration in the ANL fluidized-bed combustion flue gas system, was not completed.

The results of work completed at the University of Toronto yield no evidence that the technical objectives of the project were unobtainable. However, the project did require significantly more developmental work than was originally anticipated. Continued examination of acoustic dust conditioning for high-temperature/high-pressure applications is recommended.

TAN-JET Cyclone—A Donaldson Co. TAN-JET cyclone, claimed to have a particle removal efficiency superior to that of conventional cyclones, was installed in the ANL fluidized-bed combustor flue gas system for testing and evaluation. The TAN-JET employs a secondary flow of "clean" air through a nozzle located inside the cyclone. The Donaldson design provides for the secondary air entering the cyclone at sonic velocity.

A large number of tests have been performed with the TAN-JET cyclone to measure its particle-removal efficiency. Experiments have been performed (1) at low pressure, either at about 240 kPa with fluidization of limestone in the combustor but no coal combustion or at 305 kPa in combustion experiments, and (2) at high pressure (810 kPa) in combustion experiments.

In the lower-pressure experiments (240 and 305 kPa), loadings in the flue gas entering the TAN-JET were between about 0.5 and 3.5 g/m^3. These loadings were typical of those in gas from a primary cyclone stage of gas cleaning. At pressure drops across the secondary air nozzle of either about 105 and 140 kPa (less than that required for sonic velocity), the loadings in the flue gas leaving the TAN-JET were as low as 0.08 g/m^3 and no higher than 0.17 g/m^3. The combined particle-removal efficiency of the primary and TAN-JET cyclones in series was about 99.5% in the low-pressure combustion experiments.

In the high-pressure (810-kPa) experiments, loadings in the flue gas entering the TAN-JET ranged from about 1.0 to 6.0 g/m^3. In certain of these experiments, the primary cyclone was apparently overloaded, resulting in high inlet loadings to the TAN-JET. At these conditions, loadings in the flue gas leaving the TAN-JET were between 0.1 and 0.2 g/m^3 at the higher (105 and 140 kPa) secondary air nozzle pressure drops.

A few tests were also performed with the primary cyclone removed from the system. Loadings to the TAN-JET during these experiments were between 20 and 29 g/m^3. The TAN-JET cyclone generally exhibited a particle-removal efficiency of 99% or better at a nozzle pressure drop of 140 kPa.

Selected samples from two combustion experiments (one at 305 kPa and one at 810 kPa) were obtained for particle size analyses of the stone. The information obtained was used to determine grade efficiency curves for the TAN-JET at conditions (except pressure) that were nominally similar. Indicated 50% cut points for the TAN-JET cyclone are about 0.9 μm for the low-pressure experimental data, about 1.6 μm for the high-pressure experimental data. A vendor-supplied performance curve indicated a somewhat higher 50% cut point—about 2.0 μm.

Comparison of the loadings and size distributions of the dust passing through the TAN-JET for the above two experiments with loadings and size distributions suggested to be acceptable for gas turbines gave mixed results. In the case of the data from the 810-kPa experiment, the cumulative loading distribution by particle size generally exceeded that estimated to be acceptable. The results for the 305-kPa experiment indicated, however, that for particles larger than about 3 μm, the experimental cumulative loading by particle size fell within the range of conditions estimated to be acceptable.

Trace Pollutants in the Fluidized-Bed Combustion of Coal

The objective of this task is to develop methods of reducing the environmental impact of fluidized-bed combustion. The health and ecological effects of organic compounds and inorganic trace elements released from the FBC system will be addressed. The studies will include the identification of organic and inorganic constituents in the effluent streams (fly ash and stack gases) and

evaluation of radioactivity associated with coal combustion; identification of the FBC conditions and factors affecting combustion efficiencies and pollution; and identification of methods of removing the pollutants (*e.g.*, further catalytic burn-up, condensation/adsorption, and adsorption/desorption of vapors).

Limestone Utilization

Enhancement of Limestone Sulfation by Hydration. Laboratory Studies—The *in situ* formation of $Ca(OH)_2$ in partially sulfated "spent" limestone has been found to enhance further sulfation of the stone when the stone is reintroduced into a coal combustor. The changes in pore distribution upon dehydration (seen in combustor samples of once-hydrated ANL-9501 limestone) account for the increase in sulfation and sulfation capacity of partially spent limestones.

Laboratory experiments on fluidized-bed hydration of "spent" material have proved successful with several limestones; hydration was accomplished at low temperatures by direct spraying of water or by the introduction of steam.

Total sulfation of a variety of limestones (9) and dolomites (6) has increased dramatically after hydration. Visual inspection of the material provides a rough estimate of the extent of decrepitation induced by the expansion due to hydration.

Several general conclusions can be drawn on the basis of the results of laboratory studies, which have also been supported by examination of PDU-scale experimental samples. Increases were found in total porosity, average pore diameter, initial surface area, and particle size due to particle expansion offset by increased decrepitation. After treatment with water, total sulfation was greater in all cases.

The grain size within particles decreases as CaO is recrystallized from $Ca(OH)_2$ during dehydration. In general, upon water treatment, dolomites exhibit little or no decrepitation and there is a much smaller physical effect than for limestones. Decrepitation in limestones depends on several variables, including original porosity, the concentrations of impurities including MgO, percent $CaSO_4$, and the technique and temperature of water treatment. Precalcination following hydration increases the extent and rate of sulfation.

Several cyclic experiments have been performed on a process development unit (PDU) atmospheric fluidized-bed coal combustor. Pore distributions of the various materials from each cycle were characterized; changes in total porosity and gradual increases in average pore diameter were observed.

Differential thermal analysis (DTA) measurements were also made on these cycled samples to determine the distribution of water within the particle. Results show that almost 100% of the adsorbed water is in the form of $Ca(OH)_2$ rather than hydrated calcium sulfate.

Scanning electron microphotographs (SEMs) of laboratory and combustor samples that had been treated with water and calcined or resulfated show increased pore size and total porosity, expanded macrostructure, and decreased crystallite size of the material.

Development of Process Flowsheet for Spent Sorbent Hydration—A scheme is presented to increase the reactivity with SO_2 of partially sulfated sorbent from fluidized-bed combustion by hydrating it with water in large-scale systems.

Limestone Attrition Characterization—Natural sorbents are being tested in a laboratory-scale fluidized-bed for their attrition resistance as a function of their physical and chemical properties (for exampale, the extent of calcination or sulfation, and the predominant grain size). The objectives of this study are (1) to determine the mechanism of attrition in fluidized-beds and (2) to develop a relatively simple method of determining the attrition resistance of sorbents in fluidized-bed coal combustors.

The effect of calcination on particle size was determined for several limestones. In contrast to other stones previously tested under similar conditions, they did not break up when calcined (*i.e.*, the stones were not "poppers"). However, when some of the stones were tested for probable resistance to attrition under fluidized-bed conditions, calcination seemed to have softened the stones, causing them to lose their integrity.

In a series of experiments, Tymochtee dolomite (ANL-5101 limestone) was tested to determine the effect of attrition on the distribution of particle size in a laboratory-scale fluidized-bed (quartz vessel) combustor. Fluidized-bed attrition tests were done on two particle size ranges, 1410-1190 μm (-14 +16 mesh) and 1000-841 μm (-18 +20 mesh), for 15, 30, and 120 minutes. For the material of 1410-1190 μm particle diameter, approximately 63 wt % of the particles retained their original diameters at the end of the tests. For the particle size range, 1000-841 μm, approximately 85 wt % of the particles had the same particle diameters at the end of the tests as the original starting material. Comparison of the results for the two particle size ranges seems to show that there is a greater shift in particle diameter for the 1410-1190 μm material than for the 1000-841 μm material. However, for material of both size ranges, results suggest that after 15 minutes, the attrition rate is constant (at zero) in all test runs.

Studies of Sulfation Process in FBC—Scanning electron microphotographs have been made of limestone particles sulfated at three temperatures (800, 850, and 900°C) in the atmospheric fluidized-bed coal combustor.

At 800°C, an increased pore diameter at higher temperatures was observed, as well as incomplete calcination of the material. Characterization of combustor samples is necessary to allow their comparison with laboratory samples and ultimately an understanding of differences in reactivity of the two.

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The complete report, entitled "Support Studies in Fluidized-Bed Combustion—Annual Report, October 1978-September 1979," (Order No. PB 80 218613, Cost: $8.00, subject to change) will be available from:

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